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CHARACTERISTIC ABSORPTION OF X-RAYS: L SERIES

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Introduction.—Critical absorption wave-lengths associated with the L series of X-rays have been measured by E. Wagner¹ and M. de Broglie.² These scientists have investigated the spectra of ten of the chemical elements from tungsten to uranium, both inclusive. Each employed a photographic X-ray spectrometer with a rocksalt crystal. M. de Broglie found two critical absorption wave-lengths characteristic of each chemical element except, in the case of mercury, for which he gives only one. For bismuth, thorium and uranium he found a third, weak band.

In the investigation reported in this note we employed an ionization spectrometer with a calcite crystal. We found three critical absorption wave-lengths characteristic of each of the nine elements examined.

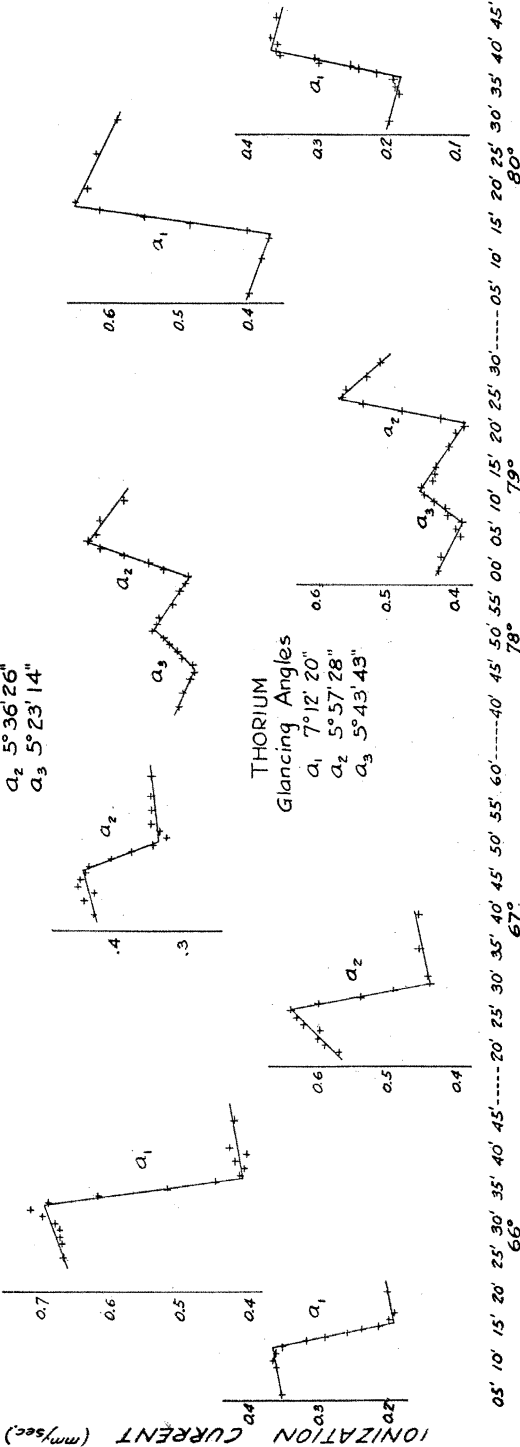
A brief discussion of the bearing of this new data on certain empirical laws and on recent theories of the mechanism of X-radiation may be found in the concluding paragraphs.

Apparatus and Method of Measurement.—A detailed description of the ionization spectrometer and the method of using it so as to obviate certain errors of measurement has been given in previous articles.³ These articles also describe the plant for generating and controlling the X-radiation. Two X-ray tubes have been employed, each equipped with a Coolidge cathode and with a thin glass window blown in a side arm. One of these tubes contained a tungsten and the other a molybdenum target. The current exciting the tubes came from a high tension storage battery. The ionization chamber contained ethyl-bromide or methyl-iodide, and sometimes a mixture of both.

In measuring a critical absorption wave-length we place a thin sheet of the chemical element to be investigated (or of a salt containing it) in the path of the beam of rays between the X-ray tube and the first slit of the spectrometer. We then measure the ionization currents for a series of positions of the crystal corresponding to wave-lengths in the neighborhood of the critical values. From these measurements we platt curves representing the ionization current as a function of the crystal table angle (see figs. 1 and 2). At a critical absorption point a sharp drop appears on the curve, indicating that X-rays of shorter wave-length than the critical value are absorbed by the chemical element to a greater extent than X-rays of longer wave-length. For the two strongest absorption bands characteristic of each element we have obtained the curves on both sides of the crystal table zero. The angular distance between the mid-points on two corresponding drops gives us twice the glancing angle, θ ,

X-RAY ABSORPTION SPECTRA
L SERIES

URANIUM
Glancing Angles
 a_1 $6^{\circ}50'30''$
 a_2 $5^{\circ}36'26''$
 a_3 $5^{\circ}23'14''$



TABLE

FIG. 1

CRYSTAL

66° 67° 78° 79° 80°
05' 10' 15' 20' 25' 30' 35' 40' 45' 50' 55' 60' 65' 70' 75' 80' 85' 90'

from which we compute the wave-length, λ , by means of the usual formula

$$\lambda = 2d \times \sin \theta,$$

where d is the distance between the reflecting planes of the crystal. The glancing angle for the third and weakest absorption band has been estimated by careful comparison with the neighboring stronger absorption drop obtained in the same series of readings.

Data Obtained.—Table 1 contains the results of our measurements. For purposes of comparison we include also the wave-lengths given by Wagner¹ and de Broglie.² The angular breadths of the drops vary from

X-RAY ABSORPTION SPECTRA L SERIES

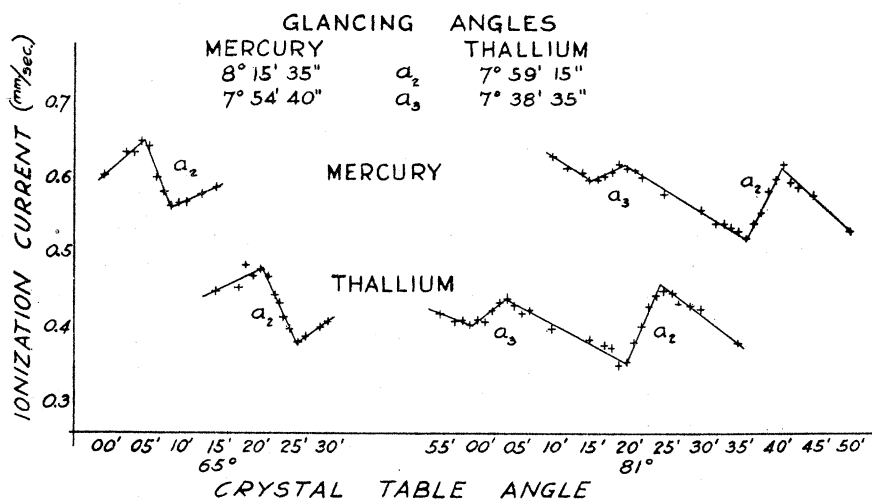


FIG. 2

three to ten minutes of arc according to circumstances, and we believe that we can estimate the positions of their centres to within ten to thirty seconds, depending upon the magnitudes of the drops and the regularity of the curves. Considerations of this kind have given us the estimates of the accuracy of each measurement recorded in the table.

Relative Positions of Emission and Absorption Wave-lengths.—In the article referred to above³ the authors have called attention to the fact that in the case of tungsten: (a) the absorption wave-length, a_3 , is slightly shorter than that of the highest frequency emission line of the L series, namely, γ_4 ; (b) the absorption wave-length, a_2 , is a large fraction of one per cent longer than that of the emission line, γ_2 ; and (c) the absorption wave-length, a_1 , is a large fraction of one per cent longer than the emission line, β_5 . The measurements on which these conclusions were based were made under the same experimental conditions for both the emission and the absorption lines. We have not measured the L series of emission

lines for chemical elements of higher atomic numbers than that of tungsten with our spectrometer. Hence we cannot make a direct comparison between emission and absorption wave-lengths obtained under the same experimental conditions for these elements. If we take the wave-lengths recorded by Siegbahn and Friman,⁴ we find that (a) our value for the absorption wave-length, La_3 , is shorter than that of the emission line, γ_4 , for each element and that the difference between the two increases slightly with the atomic number; (b) our value for the absorption wave-length,

TABLE 1
CRITICAL ABSORPTION WAVE-LENGTHS. I. SERIES OF X-RAYS
Grating Space for Calcite, $2d = (6.056 \pm 0.004) \times 10^{-8}$ cm.

CHEMICAL ELEMENT	ATOMIC NUMBER	ABSORBING SCREEN	MEASURE BY	WAVE-LENGTHS, $\lambda \times 10^3$ CM.		
				La_1	La_2	La_3
Tungsten	74		de Broglie	1.217	1.083	
Tungsten	74	Na_2WO_4	Author's	1.2136 ± 1	1.0726 ± 5	1.024 ± 3
Platinum	78		Wagner	1.072	0.934	
Platinum	78		de Broglie	1.069	0.930	
Platinum	78	Metal	Author's	1.0705 ± 3	0.9321 ± 3	0.8885 ± 9
Gold	79		Wagner	1.042	0.914	
Gold	79		de Broglie	1.038	0.898	0.858
Gold	79	Metal	Author's	1.0383 ± 3	0.8993 ± 3	0.8606 ± 8
Mercury	80		de Broglie	1.006		
Mercury	80	$HgSO_4$	Author's	1.0067 ± 5	0.8700 ± 3	0.8335 ± 9
Thallium	81		de Broglie	0.974	0.840	
Thallium	81	$Tl_2(SO_4)_3$	Author's	0.9776 ± 3	0.8415 ± 3	0.8055 ± 14
Lead	82		de Broglie	0.945	0.811	
Lead	82	$Pb(NO_3)_2$	Author's	0.9497 ± 3	0.8133 ± 3	0.7803 ± 9
Bismuth	83		de Broglie	0.921	0.786	0.753
Bismuth	83	Metal	Author's	0.9216 ± 3	0.7872 ± 3	0.7532 ± 9
		$Bi(NO_3)_3$				
Radium	88		de Broglie	0.802	0.670	
Thorium	90		de Broglie	0.757	0.624	0.604
Thorium	90	$Th(SO_4)_2$	Author's	0.7596 ± 3	0.6286 ± 3	0.6044 ± 7
Uranium	92		de Broglie	0.718	0.588	0.564
Uranium	92	Acetate	Author's	0.7214 ± 3	0.5918 ± 3	0.5685 ± 7

La_2 , is longer than that of the emission line, γ_2 , for tungsten and gold, but shorter for the other elements; (c) our value for the absorption wave-length, La_1 , is longer than that of the emission line, β_5 , for tungsten, gold, and thallium, but shorter for platinum and bismuth. The relative positions of these emission and absorption lines appear to be matters of considerable theoretical importance,⁵ and this subject will be discussed at greater length in a future note.

The Wave-lengths as Functions of the Atomic Numbers.—Many of the laws connecting X-ray wave-lengths with each other, or with the atomic numbers of the chemical elements may be expressed most simply in terms

of the wave-numbers, $1/\lambda$, or of the ratios of the frequencies of vibration to the Rydberg fundamental frequency for heavy atoms. In his classical memoirs on the wave-lengths of characteristic X-rays Moseley called attention to the fact that the square root of the wave-number (or frequency) of a line in the X-ray spectrum of an element is very nearly a linear function of its atomic number. To test this relation in the case of critical

TABLE 2
CRITICAL ABSORPTION WAVE-NUMBERS. I, SERIES OF X-RAYS
Values of $1/\lambda$ and of $\sqrt{1/\lambda}$

CHEMICAL ELEMENT	ATOMIC NUMBER	$1/\lambda$	$\sqrt{1/\lambda}$ <i>La₁</i>	DIFFERENCES
Tungsten	74	0.8240 ± 1	0.9077	
Platinum	78	0.9341 ± 3	0.9665	0.0147 ± 1
Gold	79	0.9631 ± 3	0.9814	0.0149 ± 3
Mercury	80	0.9933 ± 4	0.9966	0.0152 ± 4
Thalium	81	1.0229 ± 3	1.0114	0.0148 ± 4
Lead	82	1.0530 ± 3	1.0262	0.0148 ± 3
Bismuth	83	1.0851 ± 3	1.0417	0.0155 ± 3
Thorium	90	1.3164 ± 5	1.1473	0.0151 ± 1
Uranium	92	1.3862 ± 5	1.1774	0.0151 ± 2
<i>La₂</i>				
Tungsten	74	0.9323 ± 5	0.9656	
Platinum	78	1.0728 ± 4	1.0358	0.0176 ± 1
Gold	79	1.1120 ± 4	1.0545	0.0187 ± 4
Mercury	90	1.1494 ± 4	1.0721	0.0176 ± 4
Thalium	81	1.1883 ± 5	1.0901	0.0180 ± 5
Lead	82	1.2295 ± 5	1.1088	0.0187 ± 5
Bismuth	83	1.2703 ± 4	1.1271	0.0182 ± 5
Thorium	90	1.5908 ± 8	1.2613	0.0192 ± 1
Uranium	92	1.6898 ± 8	1.2999	0.0193 ± 3
<i>La₃</i>				
Tungsten	74	0.977 ± 3	0.988	
Platinum	78	1.1255 ± 12	1.0609	0.0182 ± 5
Gold	79	1.162 ± 1	1.0780	0.0171 ± 11
Mercury	80	1.200 ± 1	1.0953	0.0174 ± 11
Thalium	81	1.242 ± 2	1.1142	0.0189 ± 15
Lead	82	1.282 ± 2	1.1320	0.0178 ± 16
Bismuth	83	1.328 ± 2	1.1523	0.0203 ± 13
Thorium	90	1.655 ± 2	1.2863	0.0192 ± 2
Uranium	92	1.759 ± 2	1.3263	0.0200 ± 7

absorption data we have calculated the wave-numbers and the square roots of the wave-numbers for all of our values. Column 3, in Table 2, contains the wave-numbers and Column 4 their square roots. In Column 5 we have tabulated the increase in the value of $\sqrt{1/\lambda}$ per unit increase in the atomic number. It appears that these differences do not vary from

the average values of the differences in their neighborhoods by more than the estimated errors, except in one or two cases. In other words, the $\sqrt{1/\lambda}$ may be represented as a function of the atomic number by a smooth curve, from which the individual values do not differ by more than the experimental errors. In the case of the absorption band, La_1 , this curve differs a small amount from a straight line. In the cases of the bands La_2 and La_3 , however, the departures from the straight line law appear to be much greater. A very much more pronounced variation from the linear law has been noted in the case of the critical absorption frequencies associated with the K series of X-rays.⁶ These departures from the straight line law have been ascribed to the relativity change of mass with velocity of the electrons connected with the production of the X-rays.

The fact that the values of $\sqrt{1/\lambda}$ do not differ from a smooth curve law by more than the indicated errors of measurement shows that our estimates of accuracy are substantially correct.

The Relation between the Emission and Absorption Frequencies.—It has been found by experiment that the difference between the critical absorption wave-number of the K series and one of the critical absorption wave-numbers of the L series equals the wave-number of one of the α lines in the K series to within the limits of error.⁷ The experiments were performed with the X-ray spectrum of tungsten. As the emission wave-lengths in the K series have not been measured for chemical elements of higher atomic numbers than that of tungsten, we cannot test this law between the K and L series for the heavier elements. We can test the law, however, in the case of the L and M series of thorium and uranium, for here all the necessary data is available. The tables of Siegbahn and Friman⁴ contain the emission wave-lengths in the L series, and Stenström⁸ has measured three absorption wave-lengths in the M series for each of these two elements. Using our values of the L absorption wave-lengths we have compiled the following table of wave-numbers (Table 3):

TABLE 3
RELATION BETWEEN ABSORPTION AND EMISSION WAVE-NUMBERS
 L and M Series

		U	Th		U	Th		U	Th
Absorption	La_1-Ma_1	1.099	1.047	La_1-Ma_2	1.085	1.034	La_1-Ma_3	1.038	0.988
Emission	$L\alpha_1$	1.098	1.045	$L\alpha_2$	1.085	1.032			
Absorption	La_2-Ma_1	1.403	1.322	La_2-Ma_2	1.389	1.309	La_2-Ma_3	1.342	1.263
Emission	$L\beta_3$	1.408	1.319	$L\beta_1$	1.389	1.305			
Absorption	La_3-Ma_1	1.472	1.385	La_3-Ma_2	1.458	1.372	La_3-Ma_3	1.411	1.326
Emission							$L\beta_3$	1.408	1.319

It appears from this table that the wave-numbers of the emission lines $L\alpha_1$, $L\alpha_2$ and $L\beta_1$ agree very well with the differences between the absorption wave-numbers La_1-Ma_1 , La_1-Ma_2 and La_2-Ma_2 , respectively.

Judging from this data alone, there is some question as to whether the emission line $L\beta_3$ belongs to the absorption difference La_2-Ma_1 or to La_3-Ma_3 .

No emission lines have been observed in the spectrum of either thorium or uranium that correspond to the other absorption differences contained in the table.

Four emission lines have been found by Siegbahn and Friman in the spectra of both thorium and uranium in addition to those contained in our Table 3, namely, $1, \beta_2, \gamma_1$, and $\gamma_{2,3}$. The way in which the frequencies of the lines for different chemical elements vary with their atomic numbers⁴ and the researches of Webster and Clark^{9,5} on the voltages required to produce the lines in the spectrum of platinum indicate that 1 and β_2 belong to the same X-ray group as α_1 and α_2 , and therefore also to the absorption La_1 . Similarly, γ_1 belongs to the absorption La_2 . If these lines obey the frequency difference law, there must be critical absorption wave-lengths at $\lambda = 2.37 \times 10^{-8}$ cm. and at $\lambda = 2.23 \times 10^{-8}$ cm. for thorium and uranium, respectively, to correspond with the emission line 1 . These wave-lengths lie within the range of possible measurements, but the spectrum in their neighborhood has not been examined for critical absorption. Similarly, there should be critical absorption wave-lengths at $\lambda = 16.2 \times 10^{-8}$ cm. for thorium and at $\lambda = 15.8 \times 10^{-8}$ cm. for uranium to correspond with the emission lines β_2 and γ_1 . These wave-lengths, however, lie beyond the range of wave-lengths that can be measured at present by means of crystal spectrometers.

It is well known that Bohr's theory of radiation explains very simply the above relation between emission and absorption wave-numbers. The relation, however, should not be absolutely exact, if, as Sommerfeld supposes, some of the atoms contain M orbits that are elliptic, and other atoms of the same chemical element contain M orbits that are circular. In this case the L absorption drops should have a complex structure. We have not been able to observe such a structure in the drops on our curves, and the effect must be exceedingly small, if it exists at all.

Theories have been developed by Rubinowicz¹⁰ and Bohr,¹¹ according to which transfers of electrons between certain pairs of orbits cannot take place.¹² If we apply these theories to the data contained in Table 3, we find that according to both theories the transfers represented by Ma_1-La_1 and Ma_2-La_2 should occur. As a matter of fact, the corresponding lines, α_1 and β_1 , are the strongest lines in the L series. According to Rubinowicz's theory the transfer Ma_2-La_1 should occur, but according to Bohr's theory it should not, unless the atom is in a field of force. The corresponding line, α_2 , has been observed, but is relatively very faint. According to both theories the transfer Ma_1-La_1 should not occur, and, as noted above, the experiments do not indicate conclusively whether the line β_3 belongs to this combination or not. Both theories allow the transfer

Ma_3-La_1 , but no line corresponding to it has been observed. The transfer Ma_3-La_2 can occur according to Rubinowicz's theory, but not according to Bohr's, and there is no line to correspond with it. The other transfers, involving La_3 , are not covered by the theory, for it does not in its present form include a third L orbit.

Sommerfeld's L-Doublet Theory.—Sommerfeld has worked out an interesting theory of the splitting up of the L series into two distinct parts—the L series doublet. An account of his reasoning may be found in his book, *Atombau und Spektrallinien*, Chapter 5. According to this theory the difference in frequency $\Delta\nu$ between the two parts of the L series is due to the difference between the energies of electrons travelling in circular orbits and of electrons travelling in elliptic orbits. Some atoms are supposed to have circular L orbits, and other atoms of the same chemical element, elliptic L orbits. The general equation (28), in the chapter referred to, for the difference, $\Delta\nu$, in wave-number, reduces to

$$\frac{\Delta\nu}{\nu_\infty} = \frac{1}{\alpha^2}(\sqrt{2}\sqrt{1 + \sqrt{1 - b^2}} - \sqrt{4 - b^2}), \quad (1)$$

for the L doublet, where $\nu_\infty = 109737$ is Rydberg's fundamental wave-

number, $\alpha = \frac{2\pi e^2}{hc} = .007286$, $b = \alpha(N - n)$, N is the atomic number,

and n , a correction term due to the influence of neighboring electrons in the atom. Sommerfeld has tested his theory by means of the wave-number differences between certain emission lines in the spectra of a great many chemical elements, and has deduced from this data the value $n = 3.63$. Although there is some difficulty in interpreting this value of the constant, n , we have used it to test the formula by means of our values of the critical absorption wave-numbers.¹³ According to the theory the wave-number difference in the doublet should equal the difference between the wave-numbers of the absorption bands La_1 and La_2 .

TABLE 4
THE L-DOUBLET WAVE-NUMBER DIFFERENCES, $\Delta\nu \times 10^{-8}$

CHEMICAL ELEMENT	ATOMIC NUMBER	OBSERVED	CALCULATED		
			$n = 3.63$	$n = 3.45$	MODIFIED
Tungsten	74	0.1083 \pm 6	0.1071	0.1087	0.1087
Platinum	78	0.1387 \pm 7	0.1369	0.1384	0.1327
Gold	79	0.1489 \pm 7	0.1453	0.1468	0.1411
Mercury	80	0.1561 \pm 8	0.1540	0.1558	0.1496
Thalium	81	0.1654 \pm 8	0.1635	0.1652	0.1587
Lead	80	0.1765 \pm 8	0.1733	0.1751	0.1682
Bismuth	83	0.1852 \pm 7	0.1836	0.1856	0.1782
Thorium	90	0.2744 \pm 13	0.2713	0.2739	0.2639
Uranium	92	0.3086 \pm 13	0.3023	0.3044	0.2941

Table 4 contains the observed wave-number differences of the L doublet, with estimated errors of experiment, and also three sets of computed differences. The values given in Columns 4 and 5 have been calculated directly by the above formula (1) without expanding it into a series of ascending powers of b . It would be necessary to use six or more terms of such a series in order to attain the indicated accuracy.

The differences between the observed values and those computed, using Sommerfeld's value of $n = 3.63$, all have the same sign, and are distinctly greater than the errors of measurement. The values obtained with $n = 3.45$ compare more favorably with the experimental results, and indicate that with $n = 3.43$ the formula would give the observed differences, $\Delta\nu$, within the limits of error for nearly all the elements contained in the table. Even in this case there appears to be a small systematic variation between the theoretical and experimental frequency differences, as though the formula were nearly but not quite correct.

The constant n represents the repulsive force on an electron in the L orbit due to the electrons in the K orbit plus the force due to the other electrons in the L orbit. If n_1 and n_2 are the numbers of electrons in the K and the L orbits, respectively, $n = n_1 + s_{n_2}$ for circular orbits, where

$$s_n = \frac{1}{4} \sum_{s=1}^n \frac{s}{s-1} \operatorname{cosec} \frac{s\pi}{n}.$$

The values of n for elliptic orbits is assumed equal to that for circular orbits. There are, however, no values of n_1 and n_2 that agree exactly with the empirical values $n = 3.63, 3.45$ or 3.43 . For $n_1 = 2$ and $n_2 = 5$ we have $n = 3.38$. Other evidence of the distribution of electrons in atoms indicates that this arrangement is highly improbable.

As a matter of fact, the theory in its present form does not take account entirely of the influence of all the electrons in the L orbit itself. When one electron is removed from this orbit, the other electrons change their positions relative to each other and to the nucleus. In calculating the change in the atom's energy involved we must take account of the change in their energy also. A modification of Sommerfeld's formula which includes these energy changes may be obtained by taking the product of n_2 into the value of the right-hand member of equation (1), with $n = n_1 + s_{n_2}$, and subtracting from it the product of $n_2 - 1$ into the value of the same expression, with $n = n_1 + s_{n_2 - 1}$. Before computing wave-number differences from this modified formula we must assign definite values to n_1 and n_2 . This gives a sort of theoretical value for n .

Column 6, in Table 4, contains wave-number differences calculated in this way with $n_1 = 2$ and $n_2 = 4$. They differ uniformly from the experimental results by about 3%.

The above suggested alterations in the theory, however, do not obviate

certain difficulties (discussed by Sommerfeld in his book) that we encounter, if we suppose that the orbits all lie in the same plane. Possibly these difficulties will disappear, when we study more thoroughly atomic models in which the orbits do not lie in the same plane.

¹ E. Wagner, *Ann. Physik, Leipzig*, March, 1915 (868).

² M. de Broglie, *J. Physique, Paris*, May-June, 1916 (161); and *Paris C. R. Acad. Sci.*, Nov. 24, 1919 (962).

³ Blake and Duane, *Physic. Rev., Ithaca*, Dec., 1917 (624); and Duane and Patterson, *Ibid.*, 1920.

⁴ Siegbahn and Friman, *Phil. Mag., London*, April, 1916 (423); and Nov., 1916 (497).

⁵ D. L. Webster, *Proc. Nat. Acad. Sci., Washington, D. C.*, Jan., 1920 (26).

⁶ Duane and Hu, *Physic. Rev.*, June, 1918 (488); and Dec., 1919 (516); and Duane and Shimizu, *Ibid.*, Feb., 1919 (159) and Dec., 1919 (522).

⁷ Duane and Shimizu, *Ibid.*, July, 1919 (67); Duane and Patterson, *Ibid.*, 1920; Duane and Stenström, *Ibid.*, April, 1920 (302); and *Proc. Nat. Acad. Sci.*, Aug. 1902.

⁸ Stenström, *Doctor's Dissertation*, Lund, 1919.

⁹ Webster and Clark, *Proc. Nat. Acad.*, March, 1917 (181).

¹⁰ Rubinowicz, *Physik. Zs., Leipzig*, **19**, 1918 (441-465).

¹¹ Bohr, *Copenhagen Academy*, 1918.

¹² Compare Sommerfeld, *Atombau und Spektrallinien*, Chapter 6.

¹³ Compare de Broglie *Paris, C. R. Acad. Sci.*, Nov. 24, 1919 (962).

ON THE RELATIVE POSITIONS AND INTENSITIES OF LINES IN X-RAY SPECTRA

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Grouping of L Series Lines.—The wave-lengths of the lines in the *L* series of X-rays have been measured for a great many of the chemical elements by Siegbahn and Friman.¹ They have published graphs which represent the square roots of the frequencies of corresponding lines in the spectra of different elements as functions of the atomic number. The graphs for the lines 1 , α_2 , α_1 , β_2 and β_5 are very nearly straight, indicating that the square roots of the corresponding frequencies are almost linear functions of the atomic numbers, whereas the graphs for the other lines are curved, indicating a marked departure from the linear law. This would seem to mean that we can divide the lines in the *L* series into at least two groups.

In his work on characteristic absorption de Broglie² found three critical absorption wave-lengths associated with the *L* series of gold, bismuth, thorium and uranium. The authors³ have extended this, and have measured the third critical absorption wave-length as well as the other two for the elements they examined from tungsten to uranium both inclusive. The fact that three critical absorption wave-lengths appear in the *L* series of